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# Recent advances in multilayer reflective optics for EUV/x-ray sources

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**Abstract.** This paper discusses the development of (i) corrosion-resistant multilayers for the 25-80 nm region (ii) multilayer mirrors for the first 0.5-NA Micro-Exposure Tools at 13.5 nm and (iii) multilayer mirrors for the soft gamma-ray range.

## 1 Introduction

Multilayer mirrors are often essential optical elements in experiments and applications involving extreme ultraviolet (EUV) and x-ray sources. Multilayer interference coatings achieve high reflective performance due to constructive wave interference phenomena and thus enable imaging at near-normal incidence angles in the EUV/soft x-ray wavelength region. They also enable efficient operation at grazing incidence angles larger than the critical angle, thus greatly easing the fabrication, mounting and alignment of grazing-incidence x-ray optical systems. In the last few years, novel EUV/x-ray sources have emerged (4th generation synchrotrons, free-electron lasers, tabletop lasers, high-harmonic generation and attosecond sources) ushering a new era in the fields of materials science, chemistry, plasma physics, biology and life sciences. Increasingly efficient and sophisticated EUV/x-ray multilayer optics are also needed for space-borne telescopes for solar physics and astrophysics, radiation detection and medical imaging applications, high-energy physics and semiconductor photolithography.

This paper summarizes recent results and ongoing work from the development of reflective multilayer coatings for wavelengths ranging from the EUV to the soft gamma-ray regions of the spectrum. All multilayer coatings discussed below were developed and fabricated at Lawrence Livermore National Laboratory (LLNL) using DC-magnetron sputtering techniques. More details on the deposition parameters related to the multilayer coatings discussed in this paper can be found in the References provided in the following sections.

## 2 Corrosion-resistant multilayer mirrors for the 25-80 nm wavelength region

The wavelength region just below the Mg $L_{2,3}$ absorption edge (corresponding to wavelengths longer than 25 nm) contains several emission lines of highly ionized materials and coincides with the operational range of EUV sources such as tabletop lasers, high-harmonic generation sources, synchrotrons and free-electron lasers as well as solar physics studies. Mg/SiC has been shown to be the best-performing multilayer coating in the 25-80 nm wavelength region, as it possesses a unique combination of consistently high reflectivity, good spectral selectivity, thermal stability to 350 degrees C and near-zero stress[1]. However, Mg/SiC suffers from Mg-induced corrosion, an insidious problem which leads to degradation of the Mg/SiC multilayer film and its reflectivity, as shown in Fig. 1(a),(b). This problem has prevented Mg/SiC coatings from being used in applications that require long lifetime stability, which is a crucial requirement in applications such as space-borne telescopes for EUV solar physics[2]. It was established that corrosion starts from the top of the multilayer and is caused by environmental agents (reactive ions), which reach the Mg layers buried under the SiC capping (top) layer through pinholes and other defects typical in sputtered thin films. It was determined that Mg corrosion products initially appear as micron-size spots invisible to the eye, which may later grow and expand in volume, resulting in visible, eruptive effects which cause delamination of the top layers in the multilayer and exposure of the inner Mg layers.

To address this problem, Al-Mg corrosion barrier layers were developed[3]. The Al and Mg layers are deposited as two separate layers (underneath the SiC capping layer) and they spontaneously intermix to form a partially amorphous Al-Mg layer (Fig. 1(c), (d)) which provides efficient corrosion resistance while maintaining the favorable reflective properties of the original, unprotected Mg/SiC multilayer. The efficacy of this corrosion resistance concept was verified experimentally on Mg/SiC films aged for 3 years. Mg/SiC multilayers with Al-Mg corrosion barriers were demonstrated, with high narrowband reflectivity in up to 3 bands[4]. The phenomenon of spontaneous intermixing and amorphization of sputtered Al and Mg layers with nanometer-scale thickness was observed for the first time during this work. Research is now in progress to determine in detail the physics of the Al-Mg layer formation, including the timescales of spontaneous intermixing and amorphization and any crystalline phases (Al, Mg or AlMg) possibly being present in the intermixed Al-Mg layer[5].

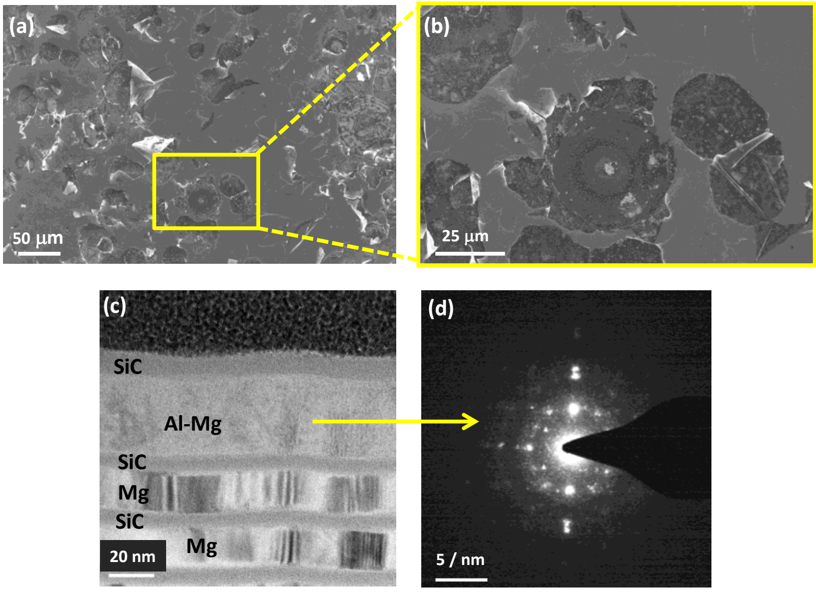


**Fig. 1 (a), (b):** Scanning Electron Microscopy (SEM) images obtained at LLNL of the top surface of a severely corroded Mg/SiC multilayer film aged for 3 years. Several areas of the coating have erupted due to the formation of voluminous Mg corrosion products, with the top portions of the coating either still partially attached to the surface, or entirely missing (from Ref. 2) **(c)** Cross-sectional TEM image of the top layers of a corrosion-resistant Mg/SiC multilayer designed for operation at wavelengths around 46 nm at normal incidence. The spontaneously intermixed, Al-Mg corrosion barrier is shown underneath the top SiC layer. (from Ref 2). **(d)** Electron diffraction image taken from the middle area of the intermixed Al-Mg layer. The appearance of diffuse rings and dispersed diffraction peaks indicates the presence of amorphous or nanocrystalline material. TEM and electron diffraction imaging were performed at Evans Analytical Group, (Sunnyvale, California).

## 3 Multilayer mirrors for the first 0.5-NA Micro-Exposure Tools at 13.5 nm

Micro-Exposure Tools (METs) are small field (30 μm × 200 μm) tools developed to provide early learning towards the extendibility of EUV lithography using 13.5 nm wavelength of illumination, especially in the areas of photoresist and mask development. METs with a numerical aperture (NA) of 0.3 have been operational for over 10 years, using laboratory (e.g: laser plasma) or synchrotron EUV sources. Next-generation METs with a NA of 0.5 (MET5) have been recently proposed[6] to demonstrate EUV patterning of features with resolution of 11 nm (half-pitch) and below. A modified

Schwarzschild design is used for the MET5 projection optics. The maximum aspherical sag and slope of the MET5 mirror substrates are a factor of 7 to 12 higher than the previous MET (NA=0.3) optics, thus presenting a significant leap in the state-of-the-art in substrate manufacturing[7,8]. Both the convex primary (M1) and concave secondary (M2) projection mirrors are rotationally symmetric aspheres. The angles of incidence (AOI) of operation in the MET5 system, defined from the normal incidence direction, range from 4 deg to 14 deg across a clear aperture radius of 14.5 to 46.1 mm for M1 and from 1 deg to 4 deg across a clear aperture radius of 39.2 to 125.4 mm for M2. The maximum height difference (sag) between the center and the edge of the reflective surface is about 3 mm for the M1 mirror and 33 mm for the M2 mirror.

The MET5 multilayer coatings are subject to extremely stringent wavefront error and wavelength matching tolerances. To minimize the wavefront error contributions of the multilayer optics in the MET5 system, the MET5 multilayer coatings were especially optimized to achieve simultaneously the highest reflectivity, lowest stress and lowest figure error (the latter being proportional to the total coating thickness). They consist of N=30 bi-layers of Mo/Si and contain a Cr under-layer that was optimized separately for each mirror. The resulting multilayer coating stress values are on the order of -100 MPa (compressive) with peak reflectance around 60%, as shown in Fig. 2. The multilayer coating profile in the lateral direction was designed for both M1 and M2 mirrors to produce a phase and a centroid wavelength (of the reflectance vs. wavelength curve) that remain constant at all locations within the mirror clear aperture, at the angles of incidence of the MET5 system. Multilayer thickness control across the curved surface of each mirror was achieved using a velocity modulation technique during deposition[9]. The non-compensable, multilayer-added figure error tolerance due to deviations from the as-designed multilayer thickness was set to 0.08 nm rms for each MET5 mirror. Fig. 3 shows the centroid wavelength obtained from EUV reflectance measurements on a multilayer-coated aspherical M2 test mirror, at the M2 angles of incidence of operation. EUV reflectance measurements were obtained at beamline 6.3.2. of the Advanced Light Source (ALS) synchrotron at Lawrence Berkeley National Laboratory. The non-compensable, multilayer-added figure error of the M2 multilayer coating was determined to be 0.022 nm rms, after subtraction of a polynomial term $ax^2+b$ which is entirely compensable via alignment shifts of the mirrors in the MET5 system. The M2 mirror centroid wavelength, calculated from the M2 profile at AOI shown in Fig. 3 and weighted by area within the clear aperture, was found to be 13.52 nm, which is within the centroid wavelength specification of 13.5 ± 0.05 nm. A more detailed discussion of the multilayer coating results from both MET5 M1 and M2 mirrors can be found in Ref. 8.

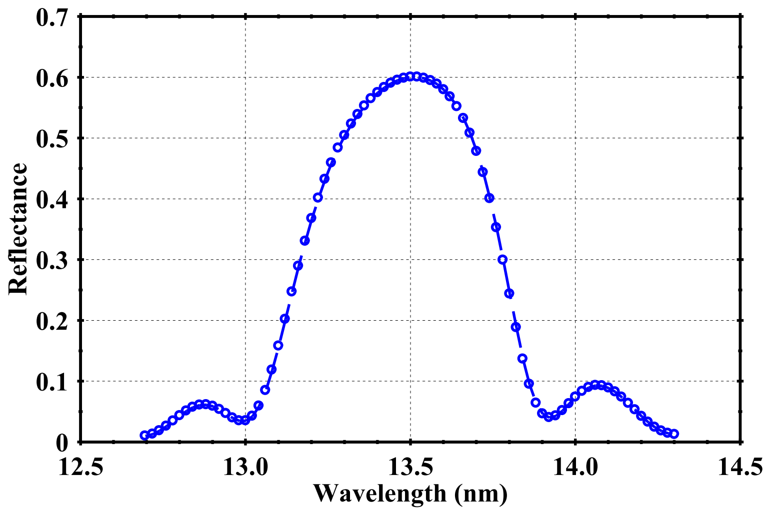


**Fig. 2:** Measured EUV reflectance of a Mo/Si multilayer with N=30 bilayers deposited on top of a Cr under-layer. The coating has 60.1% peak reflectivity and -135 MPa stress. The substrate was a Si wafer with < 0.1 nm rms high-spatial frequency roughness. This coating was developed for the M1 mirror of the MET5 system. Measurement was performed at ALS beamline 6.3.2.

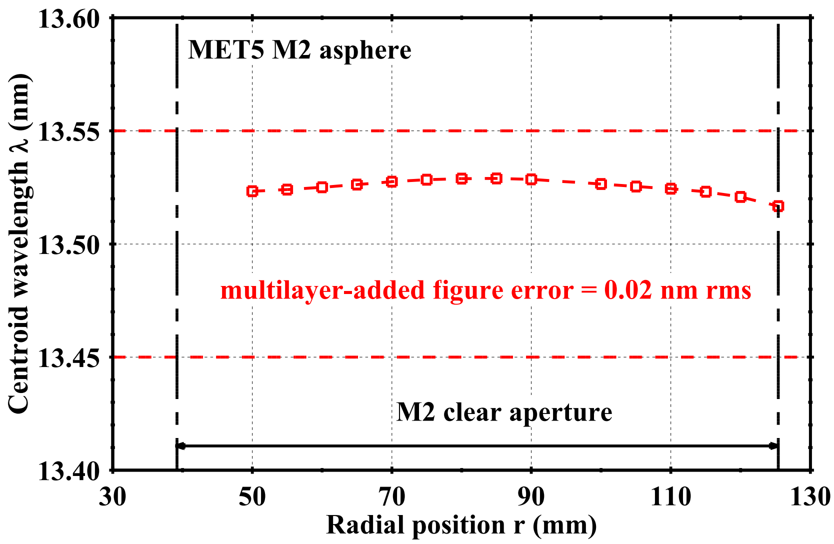


**Fig. 3:** EUV centroid wavelength measured across the clear aperture of the multilayer-coated M2 mirror at the angles of incidence (AOI) of operation in the MET system. A non-compensable multilayer-added figure error of 0.02 nm rms was determined from these results. The dash lines indicate the goal wavelength tolerance (13.5 ± 0.05 nm) at AOI. Measurements were performed at ALS beamline 6.3.2.

## 4 Multilayer mirrors for the soft gamma-ray range

In the gamma-ray photon energy range, Laue lenses (crystals) and coded apertures are typically being used as focusing or collimating elements. Multilayer-coated mirrors, if used as focusing optics operating in the soft gamma-ray, could afford large improvements in sensitivity and resolution. Efficient multilayer mirrors have recently been demonstrated experimentally at the highest reported photon energies (384, 511 and 642 keV). WC/SiC multilayer coatings with ultra-short-periods (1.5 nm) deposited on flat, super-polished glass substrates[10] were used for this purpose and achieved 52.6% peak reflectivity at 384 keV, at grazing incidence angle of 0.063 deg, measured at beamline ID15A of the European Synchrotron Radiation Facility (ESRF)[11,12].The effects of incoherent Compton scattering, which is negligible at lower photon energies but becomes significant in the soft gamma-ray, were also quantified for the first time as part of this work, using a Monte-Carlo particle simulation code. It was shown that considerable Compton scattering occurs for incidence angles in-between the Bragg resonances. Nevertheless, Compton scattering becomes insignificant for angles below the critical angle and around the Bragg reflectivity peaks, because most of the radiation is reflected by the multilayer thin film structure before it has the chance to reach the substrate and scatter[11]. These results foreshadow the use of multilayer reflective optics in soft gamma-ray applications such as nuclear[13] and medical physics and astrophysics. Of particular interest is the 511 keV photon energy corresponding to the electron-positron annihilation line, which is relevant in laser plasma diagnostics, nuclear medicine and astronomy. The ultra-short multilayer period thicknesses required for operation at gamma-ray photon energies, approach the limits of continuous layer formation and thus challenge the state-of-the-art in deposition technologies. Manipulating the deposition physics towards achieving the thinnest possible layers with the sharpest and most stable interfaces inside the multilayer structure, as needed to reach the highest reflective performance, remains an ongoing research endeavor.

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